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PATENT SPECIFICATION

(11) 1 504 172

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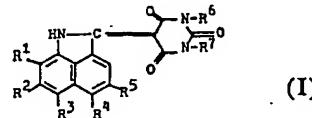
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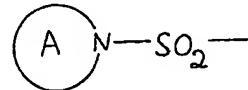
(54) NAPHTHOLACTAM DYES

(71) We, BASF AKTIENGESELLSCHAFT, a German Joint Stock Company, of 6700 Ludwigshafen, Federal Republic of Germany, do hereby declare the invention, 5 for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—
 The invention relates to dyes of the formula

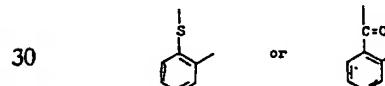
10 (I):



in which
 R¹ is hydrogen, chloro, bromo, alkyl, alkoxy, 15 nitro or arylmercapto;
 R² is hydrogen or chloro;
 R³ is hydrogen, chloro, bromo, alkyl, alkoxy, nitro, carboxylic acylamino, alkylsulfonylamino, arylsulfonylamino, alkylmercapto, 20 arylmercapto, arylsulfonyl, alkylsulfonyl, unsubstituted or substituted sulfamoyl, alkanoyl, aroyl or a heterocyclic group of the formula:—



25 in which the ring A is a saturated heterocyclic group;
 R⁴ is hydrogen, chloro, alkoxy or arylmercapto; or
 R³ and R⁴ together form a radical of the formula:



R⁵ is hydrogen, chloro or alkoxy; and
 R⁶ and R⁷ independently of one another are

alkyl; alkyl bearing hydroxy, cyano, alkoxy or carbalkoxy as a substituent; phenyl; or phenyl bearing chloro, methyl or methoxy 35 as a substituent, at least one of R¹ to R⁶ being different from hydrogen.

Interpretations of R¹ to R⁶:

It is to be understood that references herein to aryl moieties (e.g. in arylmercapto or in aryl) are intended to cover substituted 40 aryl moieties as well as unsubstituted aryl moieties.

Examples of alkyl, alkoxy and arylmercapto groups for R¹ are methyl, ethyl, methoxy, 45 ethoxy, phenylmercapto or phenylmercapto bearing chloro, methyl, methoxy, phenyl, phenoxyl or methoxycarbonyl as a substituent.

Examples for R² are the same radicals as for R¹ and also naphthylmercapto, acetyl- 50 aminophenylmercapto, acetylamino, propionylamino, benzoylamino, benzoylamino bearing chloro, methyl or methoxy as a substituent, methylsulfonylamino, ethylsulfonylamino, phenylsulfonylamino, tolylsulfonylamino, 55 methylmercapto, β -hydroxyethylmercapto, methylsulfonyl, ethylsulfonyl, phenylsulfonyl, tolylsulfonyl, chlorophenylsulfonyl, acetyl, propionyl, butyryl, benzoyl, benzoyl bearing methyl, methoxy, chloro or bromo as a substituent, sulfamoyl, N-methylsulfamoyl, N-ethylsulfamoyl, N-butylsulfamoyl, N-phenylsulfamoyl, N-chlorophenylsulfamoyl, N-methylphenylsulfamoyl, N-methoxyphenylsulfamoyl, N-trifluoromethylphenylsulfamoyl, N-methyl-N-phenylsulfamoyl, 60 N,N-dimethylsulfamoyl, N,N-diethylsulfamoyl, N,N-dipropylsulfamoyl, N,N-diethylsulfamoyl, pyrrolidinosulfonyl, piperidinosulfonyl or morpholinosulfonyl. Radicals of the formulae: NHCONH₂, NHCONHCH₃, and NHCONHC₆H₅ are also suitable,

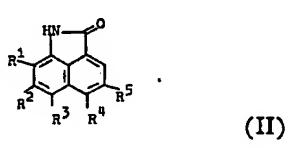
70 Examples of radicals R⁴ are hydrogen, chloro and the alkoxy and arylmercapto radicals specified for R¹.

75 Examples of alkoxy radicals R⁵ are methoxy and ethoxy.

Particular examples of R⁶ and R⁷ are alkyl of one to four carbon atoms, alkoxyalkyl of three to eight carbon atoms, hydroxyalkyl of two or three carbon atoms, cyanoethyl, alkoxy-

carbonylalkyl of one to four carbon atoms in the alkoxy and also phenyl. Specific examples are: propyl, butyl, methoxyethyl, ethoxyethyl, butoxyethyl, methoxypropyl, ethoxypropyl, pentoxypropyl, β -hydroxyethyl, β -hydroxypropyl, methoxycarbonylethyl, ethoxycarbonylethyl and butoxy carbonylethyl and preferably methyl, ethyl and phenyl.

10 Dyes of the formula (I) may be prepared by reacting a naphtholactam of the formula (II):



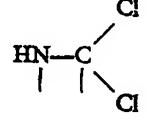
or a derivative of the same in which the grouping

$\text{HN}-\text{C}=\text{O}$

is replaced by

$\text{HN}-\text{C}-\text{S}$,
or
 $\text{N}=\text{C}=\text{S-alkyl}$,

20 $\text{N}=\text{C}-\text{O-alkyl}$,



or

$\text{N}=\text{C}-\text{Cl}$

25 with an N,N' -disubstituted barbituric acid derivative of the formula (III)



Compounds of the formula (II) are known from the literature or may be prepared by methods analogous to those described in the literature.

30 The reaction of the naphtholactams with the compounds of formula (III) proceeds in the presence of a condensing agent; when the said naphtholactam derivatives are used the presence of a condensing agent may be dispensed with.

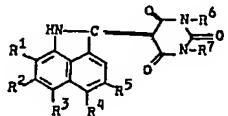
35 Phosphorus halides such as phosphorus pentachloride, phosphorus trichloride or phosphorus oxytribromide and particularly phosphorus oxytrichloride are suitable as condensing agents.

The reaction may be carried out in an inert solvent such as toluene, chlorobenzene, dichlorobenzene, nitrobenzene or dioxane or also in an excess of the condensing agent.

When the reaction is carried out with a naphtholactam derivative and without a condensing agent the abovementioned solvents are again suitable; examples of additional solvents are pyridine, glacial acetic acid, dimethylformamide and N -methylpyrrolidone.

The reactions are known in principle and details may be taken from the Examples in which parts and percentages are by weight.

The invention relates particularly to dyes of the formula:



in which

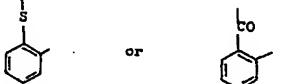
R^1 is hydrogen, chloro, bromo, C_1 to C_4 alkyl, methoxy, ethoxy, nitro, phenylmercapto or phenylmercapto bearing chloro, methyl or methoxy as a substituent;

R^2 is hydrogen or chloro;

R^3 is chloro, bromo, C_1 to C_4 alkyl, methoxy, ethoxy, nitro, acetylamino, propionylamino, benzoylamino, methylsulfonylamino, ethylsulfonylamino, phenylsulfonylamino, tolylsulfonylamino, methylmercapto, ethylmercapto, phenylmercapto, phenylmercapto bearing chloro, methyl or methoxy as a substituent, naphthylmercapto, phenylsulfonyl, phenylsulfonyl bearing chloro or methyl as a substituent, methylsulfonyl, ethylsulfonyl, N -mono- C_1 - to - C_4 -alkyl - substituted sulfamoyl, N,N -di- C_1 - to - C_4 -alkyl - substituted sulfamoyl, N -phenylsulfamoyl, N -chlorophenylsulfamoyl, N -methylphenylsulfamoyl, N -methoxyphenylsulfamoyl, N -trifluoromethylphenylsulfamoyl, pyrrolidinosulfonyl, piperidinosulfonyl, morpholinosulfonyl, C_2 to C_4 alkanoyl, benzoyl, or benzoyl bearing chloro, bromo, methyl, ethyl, methoxy or ethoxy as a substituent;

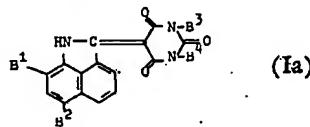
R^4 is hydrogen, chloro, methoxy, ethoxy, phenylmercapto, or phenylmercapto bearing chloro, methyl or methoxy as a substituent; or

R^5 and R^6 together are



R^5 is hydrogen, chloro, methoxy or ethoxy; and

R^6 and R^7 are independently C_1 to C_4 alkyl, C_2 to C_8 alkyl bearing hydroxy, cyano, C_1 to C_4 alkoxy or C_1 to C_4 alkoxy carbonyl as a substituent, phenyl, or phenyl bearing chlоро, methyl or methoxy as a substituent. Particular industrial importance attaches to dyes of formula (Ia):



10 in which
 B¹ is hydrogen, chloro, bromo or arylmercapto;
 B² is chloro, bromo, arylmercapto, arylsulfonyl, or aroyl; and
 B³ and B⁴ are independently methyl, ethyl or phenyl, methyl being particularly preferred.

15 The following are preferred arylmercapto, arylsulfonyl and aroyl radicals: phenylmercapto, phenylmercapto bearing chloro, methyl or methoxy as a substituent, phenylsulfonyl or benzoyl, or benzoyl or phenylsulfonyl bearing chloro or methyl as a substituent.

20

Dyes of formula (I) are yellow to violet and have high brilliance and color strength. They are suitable for dyeing synthetic fibers, particularly polyester fibers, and dyeings having very good fastness properties and particularly excellent fastness to light are obtained. Thermal resistance properties may be influenced by the choice of substituents. Dyes which sublime easily are very suitable for transfer printing.

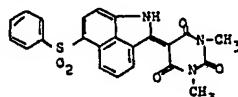
The new dyes are moreover eminently suitable for the mass coloration of plastics; fluorescent colorations having high fastness to light are obtained as a rule.

The invention includes within its scope dye formulations for dyeing synthetic fibers, the formulations containing a dye in accordance with the invention.

40 The following Examples illustrate the invention.

Example 1.

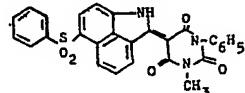
45 200 parts of phosphorus oxychloride is
 dripped into a mixture of 309 parts of 4-
 phenylsulfonylnaphtholactam - (1,8), 165
 parts of N,N' -dimethylbarbituric acid and
 1200 parts by volume of toluene at 90°C.
 The whole is stirred for 6 hours at 100°C
 and 1200 parts by volume of methanol is
 50 added during cooling so that the dye is de-
 posited in the form of brown crystals. After
 cooling, the dye is filtered off, washed with
 methanol and dried. 357 parts of the dye of
 the constitution



is obtained which dyes polyester from an aqueous liquor brilliant yellow hues of high tinctorial strength and very good light fastness and thermal stability.

Example 2.

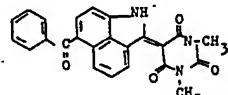
The procedure of Example 1 is followed, but the *N,N'*-dimethylbarbituric acid is replaced by an equivalent amount of *N*-methyl-*N*-phenylbarbituric acid. 427 parts of the dye of the constitution.



is obtained which dyes polyester fast golden yellow hues.

Example 3.

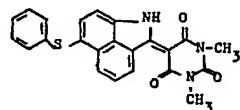
273 parts of 4 - benzoynaphtholactam - 70
 (1,8) and 190 parts of N,N'-dimethylbarbituric acid are introduced into 1000 parts by volume of toluene and stirred at 95°C. 300 parts of phosphorus oxychloride is dripped in within thirty minutes so that the starting materials pass into solution. The whole is stirred for four hours at 100°C and during cooling 2000 parts by volume of methanol is allowed to flow in. After suction filtration, washing with methanol and drying there is obtained 360 parts of the dye having the constitution: 75 80



in the form of yellowish brown crystals. The dye dyes polyester brilliant greenish yellow shades from an aqueous liquor; the dyeings have very good fastness to dry-heat pleating and setting and to light. In thermoplastics such as polystyrene fluorescent yellow hues having outstanding stability at high temperatures and outstanding fastness to light are obtained.

Example 4.

Example 4. 250 parts of phosphorus oxychloride is added at 100°C within one hour to a mixture of 277 parts of 4 - phenylmercaptanaphtholactam - (1,8), 190 parts of N,N'-dimethylbarbituric acid and 900 parts by volume of chlorobenzene. The whole is stirred for another four hours at 100°C and during cooling there is added a solution of 150 parts of triethylamine in 1500 parts by volume of ethanol. After suction filtration, washing with ethanol and drying there is obtained 349 parts of the dye of the constitution: 95 100 105



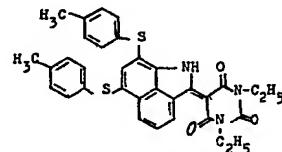
5 in the form of a dark red crystalline powder. Brilliant scarlet dyeings and prints of a high level of fastness properties are obtained on polyesters. Thermoplastics such as polystyrene for example are colored fluorescent scarlet shades having excellent stability at high temperatures and excellent fastness to light.

10 293 parts of 4 - phenylmercaptothionaphtholactam - (1,8) and 160 parts of N,N' - dimethylbarbituric acid are boiled in 400 parts by volume of N-methylpyrrolidone for eight hours. 200 parts by volume of ethanol is added during cooling. The product is suction filtered, washed and dried. 302 parts of dye is obtained which is identical with that specified in Example 4.

20 307 parts of 4 - phenylmercapto - thiophthalactam - (1,8) - S - methyl ether and 160 parts of N,N' - dimethylbarbituric acid are boiled in 600 parts by volume of acetic acid for 1 hour. The dye precipitates in the form of red crystals. Upon working up, 328 parts of dye is obtained which is identical with that specified in Example 4.

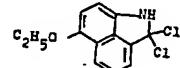
30 41.3 parts of 2,4 - bis - (4 - methylphenylmercapto) - naphthalactam - (1,8) and 20 parts of N,N' - diethylbarbituric acid are introduced into 150 parts by volume of dichlorobenzene and stirred at 95°C. 30 parts of phosphorus oxychloride is dripped in so that the components pass into solution. The whole is stirred for six hours at 95°C, diluted with 300 parts by volume of methanol and allowed to cool while stirring. The red crystals are

suction filtered, washed with methanol and dried. 49.2 parts of the dye of the constitution:



is obtained which gives in polystyrene bluish red hues of outstanding stability at high temperatures and outstanding fastness to light.

45 Example 8.
268 parts of the compound of the formula



(prepared by the method of German Laid-Open Specification 1,445,624, Example 2) is heated for 30 minutes at 100 to 120°C in 1,000 parts of xylene. 220 parts of N - methyl - N' - (3 - methoxypropyl) - barbituric acid is added and heating is continued at 100°C for 120 minutes. During cooking, 500 parts of methanol is added and the dye isolated in the usual manner. 311 parts of the dye of the constitution



50 is obtained in the form of orange crystals, which dye polyester reddish yellow hues having very good lightfastness.

55 The following dyes of the general formula (I) are prepared according to the methods specified in Examples 1 to 8.

60

65

| Ex. | R ¹ | R ² | R ³ | R ⁴ | R ⁵ | R ⁶ | R ⁷ | Hue |
|-----|--------------------------------|----------------|------------------|----------------|------------------|---|--|------------------|
| 9 | -C ₂ H ₅ | H | H | H | -CH ₃ | -CH ₃ | -CH ₃ | greenish yellow |
| 10 | H | " | -CH ₃ | " | " | " | " | " |
| 11 | " | " | Cl | " | " | " | " | reddish yellow |
| 12 | " | " | " | " | " | -C ₆ H ₅ | " | " |
| 13 | " | " | " | " | " | -C ₆ H ₅ | " | " |
| 14 | Cl | " | " | " | " | -CH ₃ | -CH ₃ | " |
| 15 | " | " | " | " | " | -C ₂ H ₅ | " | " |
| 16 | Cl | H | Cl | H | H | -CH ₃ | -(CH ₂) ₃ OCH ₃ | " |
| 17 | H | H | H | Cl | H | -CH ₃ | -CH ₃ | " |
| 18 | " | " | Cl | " | " | " | " | orange yellow |
| 19 | " | " | " | " | " | -C ₂ H ₅ | -C ₄ H ₉ | " |
| 20 | " | Cl | " | " | Cl | -CH ₃ | -CH ₃ | yellowish orange |
| 21 | " | " | " | " | " | -C ₄ H ₉ | -C ₄ H ₉ | " |
| 22 | " | " | " | " | " | -CH ₃ | -(CH ₂) ₃ OCH ₂ H ₅ | " |
| 23 | " | " | " | " | " | -C ₂ H ₅ | -C ₂ H ₅ | " |
| 24 | " | H | Br | H | H | -CH ₃ | -CH ₃ | reddish yellow |
| 25 | " | " | " | " | " | -C ₄ H ₉ | " | " |
| 26 | " | " | " | " | " | -(CH ₂) ₃ OCH ₃ | " | " |
| 27 | " | " | " | " | " | -C ₆ H ₅ | -C ₆ H ₅ | " |
| 28 | Br | " | Cl | " | " | -CH ₃ | -CH ₃ | " |

TABLE (Continued)

| Ex. | R ¹ | R ² | R' | R" | R' | R" | R ⁶ | R' | Hue |
|-----|-----------------|----------------|-------------------------------------|----|----|----|--|--------------------------------|------------------|
| 29 | Cl | H | Br | | H | H | -CH ₃ | -CH ₃ | reddish yellow |
| 30 | Br | " | " | " | " | " | " | " | yellowish orange |
| 31 | " | " | " | " | " | " | -(CH ₂) ₃ OCH ₃ | " | " |
| 32 | " | " | " | " | " | " | -(CH ₂) ₂ OCH ₂ H ₅ | " | " |
| 33 | " | " | " | " | " | " | -C ₄ H ₉ | -C ₄ H ₉ | " |
| 34 | " | " | " | " | " | " | -C ₆ H ₅ | -C ₆ H ₅ | " |
| 35 | H | H | NO ₂ | | " | " | -CH ₃ | -CH ₃ | golden yellow |
| 36 | " | " | " | " | " | " | -C ₂ H ₅ | -C ₂ H ₅ | " |
| 37 | NO ₂ | " | Br | | " | " | -CH ₃ | -CH ₃ | reddish yellow |
| 38 | Br | " | NO ₂ | | " | " | -CH ₃ | -CH ₃ | golden yellow |
| 39 | H | H | OCH ₃ | | " | " | -CH ₃ | -CH ₃ | yellow |
| 40 | " | " | " | " | " | " | -CH ₃ | -CH ₃ | " |
| 41 | " | " | " | " | " | " | -C ₆ H ₅ | -C ₆ H ₅ | " |
| 42 | " | " | OC ₂ H ₅ | | " | " | -CH ₃ | -CH ₃ | " |
| 43 | " | " | NHCONH ₂ | | " | " | -CH ₃ | -CH ₃ | " |
| 44 | " | " | " | " | " | " | -C ₆ H ₅ | -C ₆ H ₅ | " |
| 45 | " | " | NHCONICH ₃ | | " | " | -CH ₃ | -CH ₃ | " |
| 46 | " | " | NHCONHC ₆ H ₅ | | " | " | -CH ₃ | -CH ₃ | " |
| 47 | " | " | NHCOC ₆ H ₅ | | " | " | -CH ₃ | -CH ₃ | " |

TABLE (Continued)

| Ex. | R ¹ | R ² | R ³ | R ⁴ | R ⁵ | R ⁶ | R' | Hue |
|-----|----------------|----------------|---|--|---------------------------------|--------------------------------|--------------------------------|---------------|
| 48 | H | H | NHCOC ₆ H ₅ | H | H | -C ₂ H ₅ | -C ₂ H ₅ | yellow |
| 49 | " | " | NHSO ₂ C ₆ H ₅ | " | " | -CH ₃ | -CH ₃ | " |
| 50 | " | " | NHSO ₂ CH ₃ | " | " | " | " | " |
| 51 | " | " | H | -OCH ₃ | " | " | " | " |
| 52 | " | " | " | H | -OCH ₃ | " | " | " |
| 53 | " | " | " | " | -OC ₂ H ₅ | " | " | " |
| 54 | " | " | SCH ₃ | " | H | " | " | orange |
| 55 | " | " | SC ₆ H ₅ | " | " | -C ₂ H ₅ | -C ₂ H ₅ | yellowish red |
| 56 | " | " | SC ₆ H ₅ | " | " | -C ₂ H ₅ | -C ₂ H ₅ | " |
| 57 | " | " | " | " | " | -C ₂ H ₅ | -C ₂ H ₅ | " |
| 58 | " | " | " | " | " | -CH ₃ | -C ₄ H ₉ | " |
| 59 | " | " | " | " | " | -C ₆ H ₅ | -C ₆ H ₅ | " |
| 60 | " | " | Cl | SC ₆ H ₆ | " | -CH ₃ | -CH ₃ | orange yellow |
| 61 | " | " | " | " | " | -C ₂ H ₅ | -C ₂ H ₅ | " |
| 62 | " | " | SC ₆ H ₄ CH ₃ (*) | H | " | -CH ₃ | -CH ₃ | red |
| 63 | " | " | " | " | " | -C ₂ H ₅ | -C ₂ H ₅ | " |
| 64 | " | " | Cl | SC ₆ H ₄ CH ₃ (*) | " | -CH ₃ | -CH ₃ | orange |
| 65 | " | " | SC ₆ H ₄ OCH ₃ (*) | H | " | " | " | red |
| 66 | " | " | SC ₆ H ₄ OCH ₃ (*) | " | " | -C ₄ H ₉ | -C ₄ H ₉ | " |

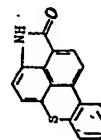
TABLE (Continued)

| Ex. | R ¹ | R ² | R ³ | R ⁴ | R ⁵ | R ⁶ | R ⁷ | Hue |
|-----|--------------------------------|----------------|---|---|----------------|--------------------------------|---|------------------|
| 67 | H | H | SC ₆ H ₄ OCH ₃ (') | H | H | -CH ₃ | -(CH ₂) ₃ OC ₂ H ₅ | red |
| 68 | " | " | Cl | SC ₆ H ₄ OCH ₃ (') | " | " | -CH ₃ | yellowish red |
| 69 | " | " | SC ₆ H ₄ Cl (') | H | " | " | " | orange |
| 70 | " | " | " | " | " | " | -C ₄ H ₉ | " |
| 71 | " | " | SC ₆ H ₃ Cl ₂ (2,5) | " | " | " | -CH ₃ | yellowish orange |
| 72 | " | " | SC ₆ H ₄ CH ₃ (') | " | " | " | " | red |
| 73 | " | " | SC ₆ H ₄ OC ₆ H ₅ (') | " | " | " | " | " |
| 74 | " | " | SC ₆ H ₄ NHCOCH ₃ (') | " | " | " | " | yellowish red |
| 75 | " | " | SC ₆ H ₄ COOCH ₃ (') | " | " | " | " | red |
| 76 | " | " | Naphthyl-2-S- | " | " | " | " | " |
| 77 | " | " | " | " | " | " | " | " |
| 78 | SC ₆ H ₅ | Cl | " | " | " | -C ₂ H ₅ | -C ₂ H ₅ | " |
| 79 | " | " | " | " | " | -CH ₃ | -CH ₃ | orange |
| 80 | " | " | NO ₂ | " | " | " | -CH ₃ | " |
| 81 | " | " | SC ₆ H ₅ | " | " | " | " | reddish orange |
| 82 | " | " | " | " | " | -C ₄ H ₉ | -C ₄ H ₉ | " |
| 83 | " | " | " | " | " | -CH ₃ | -(CH ₂) ₃ OC ₂ H ₅ | " |
| 84 | " | " | " | " | " | -C ₆ H ₅ | -C ₆ H ₅ | " |
| 85 | H | " | SC ₆ H ₅ | " | " | -CH ₃ | -CH ₃ | yellowish red |

TABLE (Continued)

| Ex. | R ¹ | R ² | R ³ | R ⁴ | R ⁵ | R ⁶ | R' | Hue |
|-----|--|----------------|----------------|--|---|----------------|--------------------------------|---|
| 86 | H | .. | H | SC ₆ H ₅ | SC ₆ H ₅ | H | -CH ₃ | yellowish red |
| 87 | SC ₆ H ₄ CH ₃ (4) | .. | H | SC ₆ H ₄ CH ₃ (4) | H | .. | -CH ₃ | red |
| 88 | .. | .. | .. | SC ₆ H ₄ CH ₃ (4) | .. | .. | -CH ₃ | -(CH ₂) ₃ OCH ₃ |
| 89 | .. | .. | .. | .. | H | .. | -C ₄ H ₉ | .. |
| 90 | SC ₆ H ₄ OCH ₃ (4) | .. | .. | SC ₆ H ₄ OCH ₃ (4) | .. | .. | -C ₄ H ₉ | .. |
| 91 | .. | .. | .. | .. | .. | .. | -CH ₃ | bluish red |
| 92 | H | .. | .. | .. | SC ₆ H ₄ OCH ₃ (4) | .. | -C ₂ H ₅ | .. |
| 93 | SC ₆ H ₄ Cl (4) | .. | .. | SC ₆ H ₄ Cl (4) | H | .. | -CH ₃ | yellowish red |
| 94 | SC ₆ H ₄ Cl (4) | .. | .. | .. | .. | .. | -C ₄ H ₉ | reddish orange |
| 95 | SC ₆ H ₄ Cl ₂ (2,5) | .. | .. | SC ₆ H ₄ Cl ₂ (2,5) | .. | .. | -CH ₃ | orange |

Example 96. 200 parts of *N,N'*-dimethylbarbituric acid and 275 parts of the compound of the following example.



is obtained in the form of a dark crystalline powder. When the dye is incorporated into polystyrene it gives violet hues having good stability to high temperature and good lightfastness properties. When the N,N-dimethylbarbituric acid is replaced by barbituric acids having other substituents, dyes having very similar hues are obtained.

Other dyes of the constitution (1) which are obtained analogously to Examples 1 to are:—

are brought into solution in 1800 parts by volume of anhydrous nitrobenzene by heating. 220 parts of phosphorus oxychloride is dripped in at 100°C and the whole is stirred for another twelve hours at 100°C. After cooling it is diluted with an equal volume of ethanol, suction filtered, washed with ethanol and dried. 321 parts of the dy's of the formula:

20

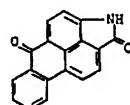
22

| Example | $R^3(R^1=R^2=R^4=R^5=H)$ | R^6 | R^7 | Hue |
|---------|---|-------------------------|---|-----------------|
| 97 | $-\text{SO}_2\text{C}_6\text{H}_5$ | $-\text{CH}_3$ | $-\text{C}_4\text{H}_9$ | golden yellow |
| 98 | „ | „ | $-(\text{CH}_2)_3\text{OC}_2\text{H}_5$ | „ „ |
| 99 | „ | $-\text{C}_4\text{H}_9$ | $-\text{C}_4\text{H}_9$ | „ „ |
| 100 | $-\text{SO}_2\text{C}_6\text{H}_4\text{CH}_3$ (4) | $-\text{CH}_3$ | $-\text{CH}_3$ | „ „ |
| 101 | „ | „ | $-\text{C}_2\text{H}_5$ | „ „ |
| 102 | „ | $-\text{C}_6\text{H}_5$ | $-\text{C}_6\text{H}_5$ | „ „ |
| 103 | $-\text{SO}_2\text{C}_6\text{H}_4\text{Cl}$ | $-\text{CH}_3$ | $-\text{CH}_3$ | „ „ |
| 104 | „ | $-\text{C}_2\text{H}_5$ | $-\text{C}_2\text{H}_5$ | „ „ |
| 105 | „ | $-\text{CH}_3$ | $-\text{C}_4\text{H}_9$ | „ „ |
| 106 | „ | $-\text{C}_4\text{H}_9$ | $-\text{C}_4\text{H}_9$ | „ „ |
| 107 | $-\text{SO}_2\text{N}(\text{CH}_3)_2$ | $-\text{CH}_3$ | $-\text{CH}_3$ | yellow |
| 108 | $-\text{SO}_2\text{N}(\text{C}_2\text{H}_5)_2$ | „ | „ | „ |
| 109 | $-\text{SO}_2\text{N}(\text{C}_4\text{H}_9)_2$ | „ | „ | „ |
| 110 | $-\text{SO}_2\text{N}(\text{C}_6\text{H}_5)_2$ | „ | „ | „ |
| 111 | „ | „ | $-\text{C}_6\text{H}_5$ | „ |
| 112 | $-\text{SO}_2\text{N}(\text{C}_6\text{H}_4\text{CH}_3)_2$ | „ | $-\text{CH}_3$ | „ |
| 113 | $-\text{SO}_2\text{N}(\text{C}_6\text{H}_4\text{O})_2$ | $-\text{C}_6\text{H}_9$ | $-\text{C}_6\text{H}_9$ | „ |
| 114 | $-\text{SO}_2\text{NHC}_6\text{H}_5$ | $-\text{CH}_3$ | $-\text{CH}_3$ | „ |
| 115 | $-\text{SO}_2\text{NHC}_6\text{H}_4\text{CH}_3$ (4) | „ | „ | „ |
| 116 | $-\text{SO}_2\text{NHC}_6\text{H}_4\text{Cl}$ (2) | „ | „ | „ |
| 117 | $-\text{SO}_2\text{NHC}_6\text{H}_4\text{OCH}_3$ (4) | „ | „ | „ |
| 118 | $-\text{SO}_2\text{NHC}_6\text{H}_4\text{CF}_3$ (3) | „ | „ | „ |
| 119 | $-\text{SO}_2\text{N}(\text{C}_6\text{H}_5)_2\text{CH}_3$ | „ | „ | „ |
| 120 | $-\text{COCH}_3$ | „ | „ | greenish yellow |
| 121 | $-\text{COC}_6\text{H}_5$ (iso) | $-\text{C}_6\text{H}_5$ | $-\text{C}_6\text{H}_5$ | „ „ |
| 122 | $-\text{COC}_6\text{H}_5$ | $-\text{C}_2\text{H}_5$ | $-\text{C}_2\text{H}_5$ | „ „ |
| 123 | „ | $-\text{CH}_3$ | $-\text{C}_4\text{H}_9$ | „ „ |
| 124 | „ | „ | $-\text{C}_6\text{H}_5$ | „ „ |
| 125 | „ | $-\text{C}_6\text{H}_5$ | „ | „ „ |

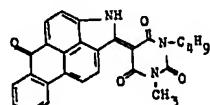
| Example | $R^3(R^1=R^2=R^4=R^5=H)$ | R^6 | R^7 | Hue |
|---------|--|-------------------------|---|-----------------|
| 126 | $-\text{COC}_6\text{H}_4\text{CH}_3$ (2) | $-\text{CH}_3$ | $-\text{CH}_3$ | yellow |
| 127 | $-\text{COC}_6\text{H}_4\text{CH}_3$ (4) | „ | „ | „ |
| 128 | „ | „ | $-\text{C}_2\text{H}_5$ | „ |
| 129 | $-\text{COC}_6\text{H}_4\text{Cl}$ (2) | „ | $-\text{CH}_3$ | greenish yellow |
| 130 | $-\text{COC}_6\text{H}_4\text{Cl}$ (4) | „ | „ | „ |
| 131 | „ | $-\text{C}_2\text{H}_5$ | $-\text{C}_2\text{H}_5$ | „ |
| 132 | „ | $-\text{CH}_3$ | $-(\text{CH}_2)_3\text{OCH}_3$ | „ |
| 133 | $-\text{COC}_6\text{H}_3\text{Cl}_2$ (2,4) | „ | $-\text{CH}_3$ | „ |
| 134 | „ | „ | $-\text{C}_4\text{H}_9$ | „ |
| 135 | $-\text{COC}_6\text{H}_3\text{Cl}_2$ (3,4) | „ | $-\text{CH}_3$ | „ |
| 136 | $-\text{COC}_6\text{H}_3\text{Cl}_2$ 2,5) | „ | „ | „ |
| 137 | „ | „ | $-(\text{CH}_2)_3\text{OC}_2\text{H}_5$ | „ |
| 138 | „ | $-\text{C}_6\text{H}_5$ | $-\text{C}_6\text{H}_5$ | „ |

Example 139.

5 208 parts of N - methyl - N' - butyl-barbituric acid and 271 parts of the compound of the constitution:



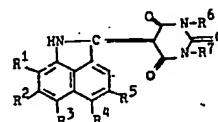
10 are stirred into 1200 parts by volume of anhydrous trichlorobenzene at 100°C. 300 parts of phosphorus oxychloride is dripped in within one hour and the whole is stirred for another eight hours at 100°C to 110°C. During cooling the whole is diluted with 800 parts by volume of methanol and then suction filtered, washed with methanol and dried. 361
15 parts of the dye of the constitution:



20 is obtained in the form of reddish brown crystals. Luminous orange colorations having good fastness properties are obtained with the dye in thermoplastics, as for example poly-styrene.

WHAT WE CLAIM IS:—

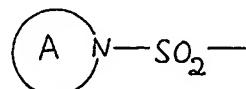
1. A naphtholactam dye of the formula:



25 in which
 R^1 is hydrogen, chloro, bromo, alkyl, alkoxy, nitro or arylmercapto;
 R^2 is hydrogen or chloro;
 R^3 is hydrogen, chloro, bromo, alkyl, alkoxy, nitro, carboxylic acylamino, alkylsulfonylamino, arylsulfonylamino, alkylmercapto, arylmercapto, arylsulfonyl, alkylsulfonyl, sulfamoyl, sulfamoyl bearing one or two substituents on the nitrogen atom, alkanoyl, aroyl or a heterocyclic group of the formula:—

30

35



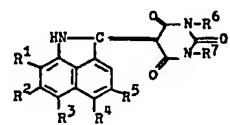
in which the ring A is a saturated heterocyclic group;
 R^4 is hydrogen, chloro, alkoxy or arylmercapto; or
 R^3 and R^4 together form a radical of the formula:—

40



5 R^6 is hydrogen, chloro or alkoxy; and
 R^6 and R^7 are independently alkyl, alkyl bearing hydroxy, cyano, alkoxy or carbalkoxy as a substituent, phenyl or phenyl bearing chloro, methyl or methoxy as a substituent; and at least one of the radicals R^1 to R^5 is other than hydrogen.

10 2. A dye of the formula:



in which

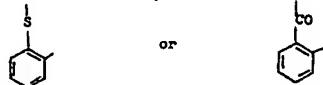
15 R^2 is hydrogen, chloro, bromo, C_1 to C_4 alkyl, methoxy, ethoxy, nitro, phenylmercapto or phenylmercapto bearing chloro, methyl or methoxy as a substituent;

R^2 is hydrogen or chloro;

20 R^3 is chloro, bromo, C_1 to C_4 alkyl, methoxy, ethoxy, nitro, acetylaminio, propionylaminio, benzoylaminio, methylsulfonylaminio, ethylsulfonylaminio, phenylsulfonylaminio, tolylsulphonylaminio, methylmercapto, ethylmercapto, phenylmercapto, phenylmercapto bearing chloro, methyl or methoxy as a substituent, naphthylmercapto, phenylsulfonyl, phenylsulfonyl bearing chloro or methyl as a substituent, methylsulfonyl, ethylsulfonyl, N - mono - (C_1 to C_8) - alkylsubstituted sulfamoyl, N,N - di - C_1 to C_8 - alkyl-substituted sulfamoyl, N - phenylsulfonyl, N - chlorophenylsulfonyl, N - methylphenylsulfonyl, N - methoxyphenylsulfonyl, N - trifluoromethylphenylsulfonyl, pyrrolidinosulfonyl, piperidinosulfonyl, morpholinosulfonyl, C_2 to C_4 alkanoyl, benzoyl, or benzoyl bearing chloro, bromo, methyl, ethyl, methoxy or ethoxy as a substituent;

35 R^4 is hydrogen, chloro, methoxy, ethoxy, phenylmercapto or phenylmercapto bearing chloro, methyl or methoxy as a substituent; or

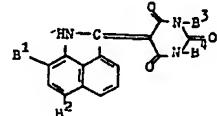
40 R^5 and R^6 together are:



45 R^6 is hydrogen, chloro, methoxy or ethoxy; and
 R^6 and R^7 are independently C_1 to C_4 alkyl,

C_1 to C_8 alkyl bearing hydroxy, cyano, C_1 to C_4 alkoxy or C_1 to C_4 alkoxy carbonyl as a substituent, phenyl or phenyl bearing chloro, methyl or methoxy as a substituent.

3. A dye as claimed in claim 1 and having the formula:



in which

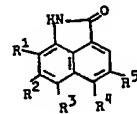
55 B^1 is hydrogen, chloro, bromo or arylmercapto;

B^2 is chloro, bromo, arylmercapto, arylsulfonyl or aroyl; and

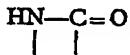
60 B^3 and B^4 independently are methyl, ethyl or phenyl.

4. A dye as claimed in claim 1 and specified in any one of the foregoing Examples.

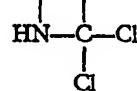
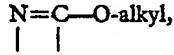
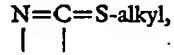
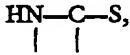
5. A process for the production of a dye as claimed in claim 1 wherein a naphtholactam of the formula:



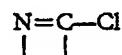
or a derivative of the same in which the grouping



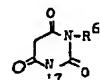
is replaced by



or



is reacted with an N,N -disubstituted barbituric acid derivative of the formula:



6. A process as claimed in claim 5 carried out substantially as described in any one of the foregoing Examples.

5 7. A dye as claimed in Claim 1 and obtained by a process as claimed in Claim 5 or Claim 6.

10 8. A dye formulation for dyeing synthetic fibers and which contain a dye as claimed in any of claims 1 to 4 or in Claim 7.

9. Plastics material whenever coloured by the presence therein of a dye as claimed in

any of claims 1 to 4 or in Claim 7.

10. Synthetic fibers which have been dyed with a dye formulation as claimed in claim 8.

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